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ABSTRACT FOR MRS 2002

Liquid de-excitation in time instead of temperature: new aspects of inhomogeneity.

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In the relaxation of hyperquenched glasses, the events normally observed during decrease of temperature (from near T_x down to T_g) can be observed isothermally as a function of time (aging). However, to reveal the entire range at one carefully chosen temperature would require measurements covering some ten orders of magnitude in time, which is inconvenient for many measurements, particularly those involving calorimetry. The projection of the glass de-excitation process onto the time axis can be speeded up in various ways that will be described. By combining differential scanning calorimetry up-scans with isothermal anneals the de-excitation process can be analyzed in considerable detail. In the present work we provide examples of this latter type of study for cases of high temperature minerals glasses that have been quenched by fibre drawing, intermediate temperature salt glasses that have been hyperquenched by twin roller quenching, and low temperature molecular glasses that have been hyperquenched by electrospraying.

An advantage of studying the system in time rather than temperature is that a partial decoupling of the vibrational excitation from the configurational excitation is achieved. For instance, the dominant anharmonic events (umklapp processes in crystals) characteristic of the higher temperature ranges of liquid behavior are now largely excluded by the very low temperatures at which the relevant configurations are studied. For the same reason, the "shapes of the basins" (referring to the configuration space energy landscape picture of Goldstein, Stillinger and Sciortino) visited by the system at the high temperatures (and in which the system is trapped during the hyperquench) can now be seen more clearly. Problems with (or subtleties in the exploration of) the single landscape paradigm are revealed.

While the primary observations to be reported will concern energy changes, observations on the vibrational density of states characterizing the system at different excitation levels will also be presented. The object is to decide the extent to which the differences in excitation rate in temperature , and the corresponding de-excitation rate in time, are controlled by changes in the vibrational density of states. These will be based on cold neutron scattering studies of (i) mineral glasses in hyperquenched, standard rate-cooled , and long-annealed states, and (ii) aqueous glassformers in mobile liquid ($T = 1.5T_g$), and glassy states.